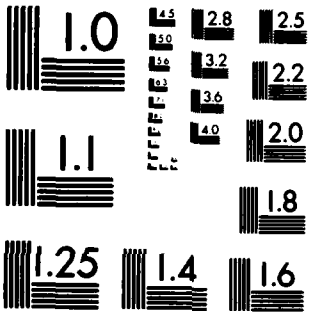
AD-A145 897 RAMAN SCATTERING FROM METASTABLE (GASB)1-XGE2X ALLOYS 1/
(U) ILLINOIS UNIV AT URBANA R BESERMAN ET AL. 1984
N00014-79-C-0424

UNCLASSIFIED F/G 7/4 NL



MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A

(1)

RAMAN SCATTERING FROM METASTABLE

$(GaSb)_{1-x}Ge_{2x}$ ALLOYS

R. Beserman*, J.E. Greene, M.V. Klein,
T.N. Krabach, T.C. McGlinn, L.T. Romano and S.I. Shah
Materials Research Laboratory and Coordinated Science Laboratory
University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

Preprint of paper to be presented at the
17th International Conference on the Physics of Semiconductors
San Francisco, California, August 6-10, 1984
Proceedings to be published by Springer-Verlag

PACS: 78.30.Gt
63.50.+x

RAMAN SCATTERING FROM METASTABLE $(GaSb)_{1-x}Ge_{2x}$ ALLOYS

R. Beserman*, J.E. Greene, M.V. Klein,
T.N. Krabach, T.C. McGlinn, L.T. Romano and S.I. Shah
Materials Research Laboratory and Coordinated Science Laboratory
University of Illinos at Urbana-Champaign, Urbana, Illinois 61801

In an extension of earlier work (1) we have used Raman Scattering to reveal structural information in mixed, metastable, crystalline ~~$(GaSb)_{1-x}Ge_{2x}$~~ $=[Ga_{1-x}Ge_x][Sb_{1-x}Ge_x]$. These alloys were deposited on GaAs substrates using a multitarget r.f. sputtering system (2).

Room temperature measurements were performed under vacuum, with the incoming laser beam perpendicular to a <100> plane, and photon polarizations along [100] directions. Figure 1 shows the Raman spectra of some of the mixed crystals and of the pure GaSb and Ge components (all normalized to same maximum peak height). When small concentrations of GaSb are introduced into Ge or small conentrations of Ge introduced into GaSb, the zone-center phonon frequency shifts, broadens, and becomes asymmetric. Figure 2 plots the frequencies versus "percent Germanium", which we define as 100x. For $x<0.1$ the GaSb-like phonon shows a TO-LO splitting, which cannot be detected for $x>0.1$. The frequency of the Ge-like optical phonon increases almost linearly with x, whereas the frequency of the GaSb-like phonon remains almost constant for $0.3 < x < 0.75$. For $x>0.75$ this mode can no-longer be detected, consistent with expectations for behavior of isolated substitutional Ga and Sb atoms in Ge: In low concentrations neither atom will give a local mode or a gap mode. Ge, however, does produce a local mode as a dilute impurity in GaSb.

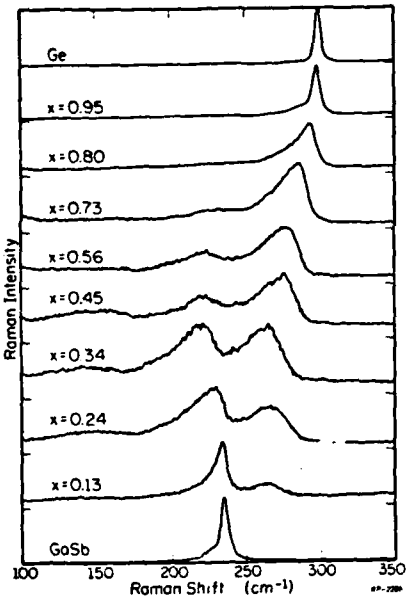


Fig. 1: Room temperature, Raman scattering of mixed $(GaSb)_{1-x}Ge_{2x}$. Scattering geometry: $z(x,y)\bar{z}$.

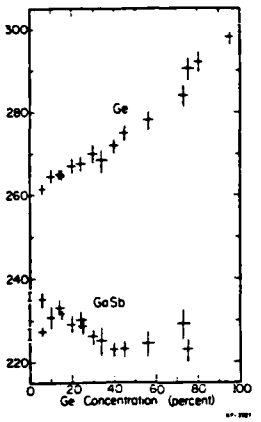


Fig. 2: Frequency dependence of peaks in $(GaSb)_{1-x}Ge_{2x}$ as a function of Ge concentration. Error bars are qualitative estimates.

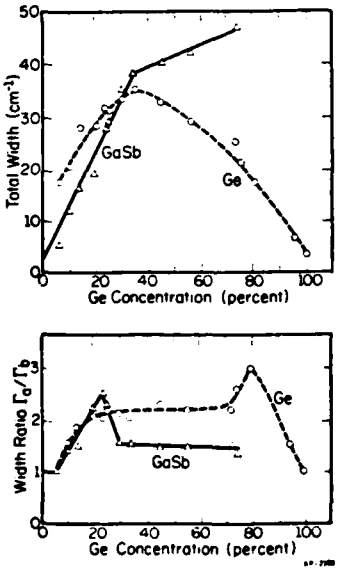


Fig. 3: Width and asymmetry at half maximum of the optical phonons.

The half widths of the optical phonons (at half-maximum) will be denoted by Γ_a and Γ_b below and above the peak, respectively. The total width $\Gamma = \Gamma_a+\Gamma_b$, and the asymmetry Γ_a/Γ_b are plotted versus x in Fig. 3. For the GaSb-like mode Γ is an increasing function of x until $x\approx0.75$, where the mode can no longer be detected. Γ for the Ge-like mode rises first linearly with (1-x), reaches a maximum for $x\approx0.35$ and then falls. The maximum values of Γ (GaSb) and Γ(Ge), 47 and 35 cm^{-1} respectively, are close to those of the pure amorphous components ($52cm^{-1}$ for a-GaSb and $47cm^{-1}$ for a-Ge). The asymmetry Γ_a/Γ_b of the GaSb-like mode increases with x until $x\approx0.25$. For $x>0.25$ Γ_a/Γ_b decreases, whereas Γ continues to increase. Similarly, the asymmetry of the Ge-like mode increases with (1-x) until $(1-x)\approx 0.20$. For $(1-x)>0.25$ the asymmetry decreases, whereas Γ continues to increase, until $(1-x)\approx0.65$. This behavior is different from that found in the quasi-binary III-V alloys $Ga_{1-x}Al_xAs$ and $Ga_{1-x}In_xAs$, where Γ_a/Γ_b is an increasing function of Γ (3) and in GaAs implanted with As^+ where Γ_a/Γ_b and Γ in samples that remain crystalline, are increasing functions of fluence (4). In these three systems Γ was never greater than about 14 cm^{-1}, and the maximum value of Γ_a/Γ_b was about 2.2.

By its very nature $[Ga_{1-x}Ge_x]$ $[Sb_{1-x}Ge_x]$ has more disorder than, say, $GaSb_{1-x}As_x$, since in the

present case both sublattices of the underlying lattice become disordered. The greater values of Γ observed here are at least partly due to this effect. An important issue is whether the non-monotonic relation of Γ_a/Γ_b to Γ is simply due to the larger amount of disorder, rather than to qualitative changes in the nature of the disorder. The abrupt changes in Γ_a/Γ_b (GaSb) near x=0.20 and Γ_a/Γ_b(Ge) near x=0.80 seem to occur over too small a change in x to be simply a quantitative effect of disorder.

In references (3) and (4) the results were analyzed using the "Spatial correlation model" which intoduces a correlation length L to account for the wave-vector relaxation (5). The contribution of phonons of finite q with frequency $\omega(q)$ and width Γ_o is accounted for by use of the following expression for the Raman line-shape:

$$I(\omega) \propto \int \exp\left(\frac{-q^2L^2}{4}\right) \frac{d^3q}{[\omega-\omega(q)]^2+[\Gamma_o/2]^2} \qquad (1)$$

$\omega(q)$ was taken to be that given by a linear chain model (3), (4). The resulting values of Γ_a, Γ_b, and Γ_a/Γ_b are increasing functions of L^{-1}. A rough extrapolation of these results to large Γ, suggests that the Γ-values in Fig. 3 imply minimum values of L of order 10Å. When a more realistic model is used for $\omega(q)$, and when both TO and LO modes are included, it is possible that for very large L^{-1} - 10^7 cm^{-1} Eq. (1) will give a line-shape with reduced asymmetry, but the change will not be an abrupt function of L^{-1}. The Γ_a/Γ_b data (Fig. 3) suggest qualitative changes in the disorder at x≈0.20-0.30 and x≈0.70-0.80. In $(GaAs)_{1-x}Ge_{2x}$, there is experimental and theoretical evidence for a transition from an average zincblende structure to an average diamond structure as x increases past $x_c \approx 0.35$ (6,7). X-ray diffraction data on $(GaSb)_{1-x}Ge_{2x}$ show a similar transition at x_c=0.30 (8). For $x>x_c$ there is no long-range zincblende order. The Raman results do not support the picture that site occupancy is completely random when $x>x_c$, because if that were the case we should see Raman scattering from Sb-Sb bonds. If these bonds were metallic, peaks should be seen at 115 cm^{-1} and 150 cm^{-1} (9). If the bonds were covalent, a peak would be seen at about 193 cm^{-1} [a frequency slightly scaled down from the Raman frequency in gray tin (10)]. There are no sharp features near 115, 150,

or 190 cm^{-1} that could be identified with Sb-Sb bonds. This fact together with the observation that the GaSb-like phonon continues to exist up to x=0.75 implies that Sb-Ga (and/or Sb-Ge) bonds exist, but not Sb-Sb bonds. If Sb-Ga bonds tend to be favored over Sb-Ge bonds, due to the coulomb interaction, there must be small regions of zincblende structure alternating with regions of anti-zincblende structure. The large values of L^{-1} suggested by the large values of Γ imply that these regions would be as small as 10 Å in extent. This figure is consistent with our failure to see any microstructure using a scanning transmission electron microscope with a resolution of 50 Å.

Whatever the nature of the short-range order or microstucture, the Γ_a/Γ_b data suggest that the structure changes at x=0.20-0.30 and again at x=0.70-0.80.

This work was supported by the Joint Services Electronic Program under N00014-79-C-0424, Office of Naval Research under N00014-80-C-0701, the National Science Foundation under the MRL grant DMR-80-20250, and the Department of Energy, Division of Materials Sciences under contract DE-AC02-76ER01198.

References

1. T.N. Krabach, N. Wada, M.V. Klein, K.C. Cadien, and J.E. Greene, Solid State Commun. 45, 895 (1983).
2. J.E. Greene, C.E. Wickersham, and T.L Zilco, J. Appl. Phys. 47, 2284 (1976).
3. P. Parayanthal, and F.H. Pollak, Phys. Rev. Lett. 52, 1822 (1984).
4. K.K. Tiong, P.M. Amirtharaj, F.H. Pollak and D.E. Aspnes, Appl. Phys. Lett. 44, 122 (1984).
5. H. Richter, Z.P. Wang, and L. Ley, Solid State Commun. 39, 625 (1981).
6. K.E. Newman, L. Lastras, B. Kramer, S.A. Barnett, M.A. Ray, J.D. Dow, J.E. Greene, and P.M. Raccah, Phys. Rev. Lett. 50, 1467 (1983).
7. K.E. Newman and J.D. Dow, Phys. Rev. B27, 7495 (1983).
8. S.I. Shah and B. Kramer, unpublished work.
9. J. Hohne, U. Wenning, H. Shultz, and S. Hufner, Z. Phys. B27, 297, (1977).
10. C.J. Buchenauer, M. Cardona, and F.H. Pollak, Phys. Rev. B3, 1243 (1970).

* Permanent Address: Solid State Institute, Physics Dept., Technion, Institute of Technology, Haifa, Israel.